AD-A267 462

Grating stimulated echo

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A theory of a grating stimulated echo (GSE) is developed. The GSE involves the sequential excitation of atoms by two counterpropagating traveling waves, a standing wave, and a third traveling wave. It is shown that the echo signal is very sensitive to small changes in atomic velocity, much more sensitive than the normal stimulated echo. Use of the GSE as a collisional probe or accelerometer is discussed.

PACS number(s): 42.50.Md, 32.80.-t

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I. INTRODUCTION

Echo phenomena are an effective means for probing relaxation processes in the field-free region between the excitation pulses. In the usual two-pulse echo [1], the signal reflects relaxation of the atomic dipole coherence, while in the stimulated echo (SE), it is atomic state populations that can be monitored. Although the SE is an effective method for studying decay rates for atomic state populations, it is not a particularly sensitive probe of relaxation associated with changes in atomic velocity. Increased sensitivity to small velocity changes can be achieved by using the grating stimulated echo (GSE) described in this paper.

The difference between the SE and GSE can be understood in terms of the Doppler phase diagram for the GSE shown in Fig. 1. The ordinate on this graph represents the Doppler phase associated with various atomic density matrix elements. A radiation pulse propagating in the $\mathbf{k}$ direction produces an atomic coherence between ground and excited states at time $T_1 = 0$. At a time T_2 before this atomic coherence has decayed away, a second pulse propagating in the $-\mathbf{k}$ direction produces ground- and excited-state population gratings. The excited-state gratings quickly decay away, but the ground-state gratings can live for a very long time (limited only by some effective ground-state lifetime). Owing to atomic motion, the ground-state gratings acquire a Doppler phase $2\mathbf{k} \cdot \mathbf{v}(t - T_2)$ in this region. At time T_3, a standing-wave pulse reverses the slope of the phase of the atomic gratings and they begin to rephase. At time $T_4 = 2T_3 - T_2$, the net grating phase is zero; a pulse propagating in the $-\mathbf{k}$ direction applied at this time once again creates an atomic state coherence. At time $t = T_4 + T_2$, the Doppler phase associated with the atomic coherence in the first and last time intervals cancel one another, and the GSE echo pulse appears.

In the "normal" SE, all pulses are copropagating and the standing-wave pulse is absent. As a consequence, no population gratings are produced and the phase evolution between times T_2 and T_4 would be represented by a horizontal line in Fig. 1.

The sensitivity of the SE and GSE to small velocity changed δv in the time interval $(T_4 - T_2)$ is dramatically different. In the SE, the velocity changes modify the per-

fect phase cancellation of the atomic state coherence only. This phase change is of order $|\mathbf{k} \cdot \delta v T_2| \leq |\mathbf{k} \cdot \delta v / \gamma|$, where γ is the rate at which the atomic coherence decays. On the other hand, owing to the Doppler phase associated with the population gratings, the GSE's sensitivity to velocity changes is of order $|\mathbf{k} \cdot \delta v (T_4 - T_2)| \gg |\mathbf{k} \cdot \delta v T_2|$. The production of population gratings in echo phenomena
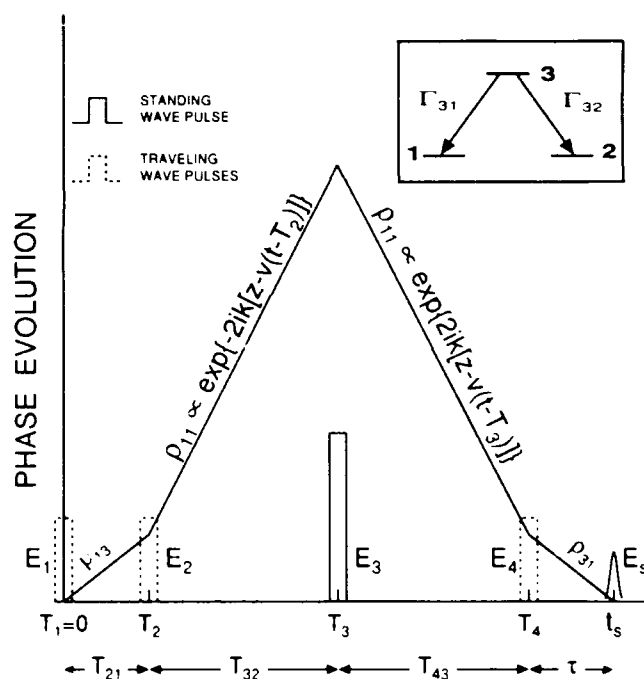

FIG. 1. Doppler phase associated with various density matrix elements leading to a grating stimulated echo (GSE). The four input pulses E_i ($i = 1-4$) give rise to an echo E_s at $t = t_s$. The field propagation vectors are $\mathbf{k}_1 = -\mathbf{k}_2 = -\mathbf{k}_4 = \mathbf{k}_s$. There is a second contribution to the echo signal (not shown) for which $\mathbf{k}_4 = \mathbf{k}_1 = -\mathbf{k}_s$; that contribution is obtained by reflection about the t axis and involves density matrix elements $\rho_{11}, \rho_{11}, \rho_{11}, \rho_{11}$. The corresponding diagram for the stimulated echo (SE) has $E_3 = 0$ and $\mathbf{k}_1 = \mathbf{k}_2 = \mathbf{k}_4 = \mathbf{k}_s$; the Doppler phase for the SE is constant in the time interval T_{42}. The atomic energy-level diagram is shown in the inset. Each level decays to an external reservoir at rate γ_l and the overall decay rate of level 3 is $\Gamma = \Gamma_{31} + \Gamma_{32} + \gamma_l$. The incident fields drive only the 1-3 transition.

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was first discussed by Mossberg *et al.* [2].

In Sec. II, an expression for the GSE is derived. The long-lived ground-state grating exists only if the sum of ground- plus excited-state population is not conserved [3,4]; consequently, the atomic model we choose is one in which the ground state consists of two sublevels. In Sec. III, two applications of the GSE are discussed. It is shown that the GSE can be used as a sensitive probe of collisions occurring between ground-state "active" atoms and perturbers. Furthermore, it is shown that the GSE can serve as the basis for an atom accelerometer. Such an accelerometer is similar to those proposed by Borde [5] and demonstrated experimentally by Riehle *et al.* [6] and Kasevich and Chu [7]. An essential distinction is that the GSE does not require a long-lived excited state, as in [6], and is not connected with the establishment of a coherence between ground-state sublevels, as in [7].

II. ECHO FORMATION

Let us consider a gas of three-level atoms interacting with a radiation field $E(r,t)$. The upper atomic level 3 decays to level 1 at rate Γ_{31} and to level 2 at rate Γ_{32} (see Fig. 1). The field

$$E(r,t) = \tfrac{1}{2}\hat{z}e^{-i\Omega t}\sum_{j=1}^{4} E_j g_j(t-T_j)$$

$$\times \begin{Bmatrix} e^{ik_j x}, & j=1,2,4 \\ \cos(kx), & j=3 \end{Bmatrix} + \text{c.c.},\qquad (1)$$

consists of a number of pulses, each having central frequency Ω. Pulse j ($j=1$–4) has amplitude E_j, and is characterized by a pulse envelope function $g_j(t-T_j)$ centered at $t=T_j$, having a temporal width of order τ_j. Pulse intervals T_{ik} are defined by $T_{ik}=T_i-T_k$ and T_1 is set equal to zero. All pulses are polarized along the z axis. The propagation directions are chosen as $k_1=-k_2=k$ and $k_4=\pm k$, where $k=\Omega/c$.

We wish to calculate the echo signal generated at times $t>T_4$. The master equation for the density matrix ρ (in the interaction representation) is

$$\left[\frac{d}{dt}+\Gamma\right]\rho_{33}=-(i/\hbar)[V,\rho]_{33},\qquad (2a)$$

$$\left[\frac{d}{dt}+\gamma_l\right]\rho_{11}=\Gamma_{31}\rho_{33}-(i/\hbar)[V,\rho]_{11}+\gamma_l W_1(v),\qquad (2b)$$

$$\left[\frac{d}{dt}+\gamma_l\right]\rho_{22}=\Gamma_{32}\rho_{33}+\gamma_l W_2(v),\qquad (2c)$$

$$\left[\frac{d}{dt}+\gamma\right]\rho_{13}=-(i/\hbar)[V,\rho]_{13},\qquad (2d)$$

where $d/dt=\partial/\partial t+v\partial/\partial x$, v is the x component of the atomic velocity, $V=-\hat{d}\cdot E(r,t)$ is the atom-field interaction Hamiltonian, $\hat{d}$ is the atomic dipole moment operator, $\Gamma=\Gamma_{31}+\Gamma_{32}+\gamma_l$ is the upper level decay rate, $\gamma_l\ll\Gamma$ is some state-independent, effective loss rate for the atoms, $\gamma\approx\Gamma/2$ is the rate at which the coherence ρ_{13} decays, and $W_i(v)$ ($i=1,2$) is the level i equilibrium velocity distribution in the absence of the fields.

Equations (2) are solved assuming that $|\Delta\tau_j|\ll1$, $\Gamma\tau_j\ll1$, $ku\tau_j\ll1$, where $\Delta=\Omega-\omega_{31}$ is the detuning of the field frequency relative to that of the atomic transition $3\rightarrow1$ and u is the most probable atomic speed. Furthermore, the traveling-wave pulses are treated to lowest order in perturbation theory. These restrictions are imposed for mathematical simplicity, they are in no way critical to the effect to be derived herein.

It is convenient to carry out the calculation in the atomic rest frame. The transformation between the laboratory and atomic frames is given by $x_0=x-vt$, where x_0 is the atomic position at $t=0$. Following a rather straightforward calculation [1,8,9], one arrives at the density matrix for $t>T_4$. Keeping only that part of the density matrix responsible for echo formation at $t=t_s$ (see Fig. 1), one finds

$$\rho(t)=(i/16)(\Gamma_{32}/\Gamma)^2\exp[-\gamma(\tau+T_{21})-\gamma_l T_{42}]W_1(v)\theta_4$$

$$\times\exp[ik_4(x_0+vT_4)-i\Delta T_4]\{\theta_2\theta_1^*\exp[i(k_2-k_1)x_0-i(\Delta-k_2v)T_{21}]+\text{c.c.}\}$$

$$\times\cos\{|\theta_3|\cos[k(x_0+vT_3)]\}|3\rangle\langle1|+\text{H.c.}\qquad (3)$$

where $\theta_j=-(d^*E_j/\hbar)\int_{-\infty}^{\infty}dt\,g_j(t)$ ($|\theta_j|$ is the area of pulse j with $|\theta_j|\ll1$ for $j=1,2,4$), $d=\langle1|d_z|3\rangle$ and $\tau=t-T_4$.

For an echo signal having propagation vector $k_s=\pm k\hat{x}\equiv k_s\hat{x}$, the positive frequency part of the echo amplitude $E_s(t)=2\pi ikLP_s(t)$ is proportional to the positive frequency part of the k_s spatial harmonic of the polarization $P_s(t)$ which, in turn, is related to the atomic density matrix via

$$P_s(t)=nde^{i\Delta\tau}(1/L)\int_0^L dx\,e^{-ik_s x}\int_{-\infty}^{\infty}dv\langle3|\rho(t)|1\rangle,\qquad (4)$$

where L is the length of the gas cell along the x axis and n is the atomic density. Substituting expression (3) into (4), setting $x=x_0+vt$, and using the fact that $\cos(a\cos\phi)=\sum_m(-1)^m J_{2m}(a)e^{2im\phi}$, where $J_q(a)$ is the Bessel function of order q, one finds the positive frequency part of the polarization is

$$P_s(t)=(i/16)nd(\Gamma_{32}/\Gamma)^2\exp[i\Delta\tau-\gamma(T_{21}+\tau)-\gamma_l T_{42}](\theta_4\theta_2\theta_1^* e^{-i\Delta T_{42}v}p+\theta_4\theta_2^*\theta_1 e^{i\Delta T_{42}v}p'),\qquad (5a)$$

where

$$P = \frac{k}{2\pi} \sum_{m=-\infty}^{\infty} (-1)^m J_{2m}(|\theta_3|) \int_0^L dx_0$$

$$\times \int_{-\infty}^{\infty} dv\, W_1(v) \exp\{ix_0(k_4+k_2-k_1+2mk-k_s) + iv[T_{32}(k_4+2mk-k_s) + T_{43}(k_4-k_s)]$$

$$+ iv[T_{21}(k_4+k_2+2mk-k_s) - k_s\tau]\},$$ (5b)

and P' is obtained from (5b) with the replacements $k_1 \to -k_1$, $k_2 \to -k_2$.

Let us consider expression (5b) for P. The spatial phase must vanish for a coherent contribution to the echo field. This condition, together with the fact that $k_1 = -k_2 = k$, leads to a value for the propagation constant, $k_s = k_4 + 2(m-1)k$. Since $kuT_{32} \gg 1$ and $T_{32} \gg T_{21}$ it is not possible to get a nonvanishing result after averaging over v unless $T_{43} \approx T_{32}/(m-1)$. Integral values $m \geq 2$ are allowed. It then follows that the phase-matching condition $[k_s = \Omega/c]$ can be satisfied only for $m = 2$ ($T_{43} = T_{32}$), $k_4 = -k$, and $k_s = k$ (echo propagation in the $+x$ direction). In this case, the echo amplitude is

$$E_s(t) = -(\pi kL/8)nd(\Gamma_{32}/\Gamma)^2\theta_4\theta_2\theta_1^* \exp[-\gamma(T_{21}+\tau) - 2\gamma_l T_{32} + i\Delta(\tau-T_{21})]J_4(|\theta_3|)F_1(\tau-T_{43}),$$ (6)

where $F_1(t) = \int_{-\infty}^{\infty} dv\, W_1(v)e^{-ikvt}$.

The corresponding result for the P' term of Eq. (5a) is $T_{43} = T_{32}$, $k_4 = k$, and $k_s = -k$. In this case, the echo signal propagates in the $-\hat{x}$ direction, having amplitude $E_s'(t) = \theta_4[E_s(t)/\theta_4]^* \exp(2i\Delta\tau)$.

III. DISCUSSION OF THE ECHO SIGNAL: APPLICATIONS

(1) The structure of the echo depends on the homogeneous width γ and inhomogeneous width ku. Two limiting cases of experimental interest are $ku \gg \gamma$ and $ku \ll \gamma$. The first condition is typical for atoms at room temperature while the latter typical for sub-Doppler, laser cooled atoms.

For $ku \gg \gamma$ one finds that the GSE has a temporal width of order $(ku)^{-1}$ and is centered at $t_s = T_4 + T_{21}$, assuming $T_{43} = T_{32}$. The backward-directed echo contains Ramsey fringe structure, owing to a factor of $\exp(2i\Delta T_{21})$ in the echo amplitude, which is typical for echoes induced by the separated counterpropagating waves [10]. Such a factor is absent for the forward directed echo; however, it is possible to create Ramsey fringes for the forward-directed echo by introducing a difference $\delta T = T_{43} - T_{32}$ between the pulse intervals. If $\delta T \leq \gamma^{-1}$, one does not destroy the echo signal, which is centered at $t_s = T_4 + T_{21} - 2\delta T$. The forward directed echo amplitude contains a factor of $\exp[2i\Delta(\delta T)]$ which gives rise to Ramsey fringe structure.

When $ku \ll \gamma$, the time scale of the echo experiment must be much larger than γ^{-1} to allow for inhomogeneous relaxation of the different atomic velocity subgroups. Consequently, the standard two-pulse excitation scheme (two pulses separated by a time interval of order γ^{-1}) [1] does not lead to echo formation. Excitation pulse sequences similar to those of the SE or GSE, with $T_{42} \gg \gamma^{-1}$, are needed for echo formation in the limit $ku \ll \gamma$. The temporal structure of the echo is very different than that for $ku \gg \gamma$. When $ku \ll \gamma$, the echo signal appears as soon as the pulse at $t = T_4$ is applied, and has a duration of order γ^{-1}. In the SE, the echo propagates in the same direction as both the last excitation pulse and the free decay signal following this pulse.

Only the specific dependence on the delay T_{32} would permit one to extract the SE contribution to the total signal. In contrast, the GSE propagates in a direction opposite to that of the last excitation pulse and can be spatially separated from both the driving pulse and the free decay signal. Thus the GSE offers a distinct advantage over the SE for the observation of such a signal.

(2) Aside from the directionality properties, the major advantage of the GSE over the SE is its sensitivity to small velocity changes. The GSE echo amplitude, $E(\text{GSE})$, is proportional to (see Fig. 1) $\exp[ikv(T_{21}-\tau)]\exp[2ikv(T_{43}-T_{32})]$, while the corresponding SE amplitude, $E(\text{SE})$, (with $k_1 = k_2 = k_4 = k$ and $k_3 = 0$) is proportional to $\exp[ikv(T_{21}-\tau)]$, reflecting the fact that the population part of the phase diagram (Fig. 1) is horizontal between T_2 and T_4 for the SE. Any small change δv in v results in a phase shift of order $2k\delta v(T_{21}+T_{32})$ for the GSE, but only $k\delta v T_{21}$ for the SE. Since $T_{32} \gg T_{21}$, the sensitivity to small δv is much larger for the GSE than the SE. This feature has been exploited by Kasevich and Chu in constructing an atom interferometer based on the Raman effect [7].

The sensitivity to small δv makes the GSE an ideal tool for probing atomic collisions. Consider the limiting case when $k\delta v T_{21} \ll 1$ and $\Gamma_{13}T_{12} \ll 1$, where Γ_{13} is the collision rate associated with the atomic coherence ρ_{13}. Assuming, moreover, that $\Gamma T_{32} \gg 1$, $\Gamma_i' \ll \Gamma$ (Γ_i' is the collision rate for atoms in state $i = 1,2,3$) and that the collision kernel $W_i(v',v)$ ($i = 1,2,3$), defined as the probability density per unit time that an atom in state i changes its velocity from v' to v, can be written as $W_i(v',v) = \Gamma_i' f_i(v-v')$ [11], where $f_i(\epsilon) = f_i(-\epsilon)$ and $\int f_i(\epsilon)d\epsilon = 1$, one can show that the SE amplitude is given by [12]

$$E(\text{SE}) \propto A(\text{SE})\exp(-2\gamma_l T)$$

$$\times \exp(-\Gamma_l' T k^2 \langle\delta v^2\rangle_l T_{21}^2),$$ (7)

where

$$A(\text{SE}) = \frac{\Gamma_{32}}{\Gamma} + \frac{\Gamma_{31}}{\Gamma^2}\frac{k^2 T_{21}^2}{2}(\Gamma_3'\langle\delta v^2\rangle_3 - \Gamma_1'\langle\delta v^2\rangle_1),$$

$\langle\delta v^2\rangle_i = \int \epsilon^2 f_i(\epsilon)d\epsilon$, and $T = T_{32} = T_{43}$. The corresponding expression for the GSE is

$$E(\text{GSE}) \propto A(\text{SE})\{(\Gamma_{32}/\Gamma) + (\Gamma_{31}/\Gamma^2)[A_3(2kT) - A_1(2kT)]\}\exp(-2\gamma_l T)\exp\{-2\Gamma_l^* T[1 - H_1(T)]\}, \tag{8}$$

where $A_i(y) = \Gamma_i^*[1 - \int f_i(\epsilon)\exp(-iy\epsilon)d\epsilon]$ and $H_i(t) = t^{-1}\int_0^t dt'\int d\epsilon f_i(\epsilon)\exp(2ik\epsilon t')$. Note that, if $\Gamma_1^* = \Gamma_3^*$, $f_1(\epsilon) = f_3(\epsilon)$, and $\Gamma_{32} = 0$, population is conserved for each velocity subgroup (except for an overall decay rate) and the SE and GSE amplitudes vanish [4].

In comparing the SE and GSE signals, one notes the following differences: (1) The SE amplitude depends only $\langle \delta v^2 \rangle_i$ and not on the specific form of the kernel, while the GSE amplitude could, in principle, be used to extract the form of $f_1(\epsilon)$. (2) The minimum velocity change in a single collision detectable using the GSE is of order $(2kT)^{-1} \geq \gamma_l/2k$, whereas it is $(2T/T_{21}) \gg 1$ times larger for the SE. (3) The minimum collision rate Γ_1^* detectable using the GSE is of order $(2T)^{-1} \geq \gamma_l/2$, whereas it $2(k^2\langle \delta v^2 \rangle_i T_{21}^2)^{-1} \gg 1$ times larger for the SE. Thus, the GSE is a much more sensitive probe of collisional effects than the SE.

For thermal samples, one can take $\gamma_l \simeq 10^4$ s^{-1} to simulate some effective lifetime of the atoms in the atom-field interaction region. In that case, one can use the GSE to measure δv's as small as 0.1 cm/s (taking $k = 10^5$ cm^{-1}) and collision rates $\Gamma_1^*/2\pi$ as small as 10^3 Hz [13]. For cold atoms the sensitivity increases linearly with the increased atomic lifetime in the interaction zone.

(3) Inertial effects can significantly modify the phase of the GSE. We calculate the effect of a gravitational acceleration along the $\mathbf{k}$ direction on the amplitude of the GSE, taking into account the temporal phases ϕ_1, ϕ_2, ϕ_3, and ϕ_4 of the excitation pulses. We find that the phase of the echo signal ϕ_s can be expressed as

$$\phi_s - \phi_4 = \pm[(\phi_2 - \phi_1) - \tfrac{1}{2}kgT_{42}^2] \tag{9}$$

where g is the acceleration of gravity and $+$ and $-$ refer to the P and P' terms, respectively. We have ignored terms of order kgT_{21}^2 and $kgT_{21}T_{43}$. The phase ϕ_s may be measured by a homodyne technique using a reference laser as both the source of the excitation pulses and as a local oscillator for the echo phase detection. It is clear from Eq. (9) that the reference laser need be phase coherent only over a time of order T_{21}, and not over the duration of the complete echo sequence T_{42}. Since, according to Eq. (9), T_{21} may be made arbitrarily short without loss of sensitivity to g, a highly stabilized laser is not required for the measurement. By comparison, the gravitational phase shift for the SE is $\phi_s - \phi_4 = \pm[(\phi_2 - \phi_1) - kgT_{42}T_{21}]$, smaller than that of the GSE by a factor of $T_{21}/T_{42} \lesssim 1/\gamma T_{42}$. To achieve high precision with the SE, a long-lived excited state is required as well as a very stable laser source.

As an example, we calculate the sensitivity to gravitational acceleration of the GSE applied to a gas of atoms at room temperature. Assuming an uncertainty in the phase measurement of $\delta\phi_s$, the corresponding relative uncertainty in a measurement of g is $\delta g/g = 2\delta\phi_s/kgT_{42}^2$. For $k \sim 10^5$ cm^{-1} and $T_{42} \sim 10^{-4}$ s (limited by the atoms' lifetime in the interaction volume), one finds $\delta g/g \sim 2\delta\phi_s$. Thus, for a phase sensitivity of 10^{-3}, one could measure the gravitational force on a gas of room-temperature atoms to a precision of about 0.1%. For a gas of laser cooled atoms, one should be able to achieve the same sensitivity δg as that reported in Ref. [7].

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